A COMPUTER PROGRAM TO FACILITATE PERFORMANCE ASSESSMENT OF UNDERGROUND LOW-LEVEL WASTE CONCRETE VAULTS

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Abstract

A computer program (4SIGHT) to facilitate performance assessment of underground concrete vaults for low level waste (LLW) disposal facilities is being developed at the National Institute of Standards and Technology (NIST). Specifically, the program predicts the hydraulic conductivity and the service life of an underground concrete vault. The hydraulic conductivity estimate is based upon empirical relations. The service life is estimated from consideration of three major degradation processes: steel reinforcement corrosion, sulfate attack, and leaching. The performance prediction is based upon ion transport equations for both diffusion and advection. Most importantly, the computer program incorporates the synergistic degradation effects of all three processes, and their effect upon the transport coefficients.

Introduction

The performance analysis of underground low-level waste (LLW) disposal facilities is a complex task. A portion of this task consists of modelling ground water flow surrounding the buried vault. Completing this calculation requires knowledge of the hydraulic conductivity (permeability) and the service life of the vault, which will likely be constructed of concrete. Published data could be used to estimate the hydraulic conductivity, but various degradation processes must be considered when estimating the service life of the concrete.

Mechanistic models exist for predicting service performance of concrete due to individual degradation mechanisms. Unfortunately, an analytic solution to simultaneous degradation processes controlled by diffusion, Darcy flow, and chemical reactions in the pore space, is virtually impossible. However, since all of the degradation processes considered here depend upon the movement of ions, a computer program can be written which predicts the ion transport through the vault walls, maintains chemical equilibrium, and calculates precipitation/dissolution of salts. This is the approach taken in developing the computer program 4SIGHT [1].

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Model

Simulating advective and diffusive transport throughout an entire LLW vault is an ambitious task; the vault is a complex structure and virtually every vault will be unique. However, the problem can be simplified greatly by realizing that the critical element of the vault is the roof because it is the element most likely to be in a moist environment, which facilitates damage due to ion transport.

The analysis can be simplified further by considering the transport through a slab which approximates a roof. Due to the symmetry of this reduced problem, an estimate of the service performance of a vault can be simplified to a one-dimensional analysis of the flow through the center of a slab. 4SIGHT implements this analysis using a one-dimensional finite difference routine which calculates ion transport. The program is composed of a linear array of computational elements, each of which incorporate the transport properties of the concrete, and contain the concentration of diffusing ion species and the quantity of solid salts present.

Degradation

4SIGHT incorporates the main degradation processes typically found in concrete structures: corrosion of steel reinforcement, sulfate attack, and leaching. A detailed discussion of these processes can be found in Clifton and Knab[2]. The effects of cracks upon the transport properties are incorporated, though they are not a degradation process *per se*. Also, the effects of freezing and thawing cycles were not considered because no reliable model exists. This omission from the analysis will likely have very little impact since most vaults will be buried below the frost zone.

Corrosion of steel reinforcement

Normally, the pore solution of concrete is alkaline (pH $\approx$ 13), which passivates steel, inhibiting corrosion. Under this condition, the corrosion rate is considered negligible because insufficient data exist to quantify the rate of corrosion in alkaline concrete pore solutions absent of Cl^-ions. As $Cl^-$and other anions are transported into the concrete, the pH decreases. When the mass of Cl^-at the reinforcement reaches 0.04% of the concrete mass, the passivated layer around the reinforcement is neutralized, initiating corrosion. For good quality concrete, the interval of time between initiation of corrosion and structural failure is far smaller than the time required for the $Cl^-$to diffuse to the reinforcement in sufficient quantity to initiate corrosion. Therefore, the time of failure is approximated by the time required for the Cl^-concentration at the reinforcement to achieve a sufficient level of concentration to initiate corrosion[2].

Sulfate attack

No accurate physicochemical model exists for sulfate attack. Lacking this, the computer program employs the linear attack rate model of Atkinson and Hearne[3]:

$$R = \frac{E\beta^2 c_o C_E D}{\alpha\gamma(1-\nu)} \tag{1}$$

where R is the velocity of sulfate front, E is the Youngs modulus of the concrete, β is the linear strain due to one mole of Na^+per m^3, c_o is the external SO_4^{2-}concentration, C_E is the concentration of reacted sulfate as ettringite, D is the diffusion coefficient of SO_4^{2-}, α is the roughness factor, γ is the fracture surface energy, and ν is the Poisson ratio. The report by Atkinson and Hearne give typical values for each parameter. Note that the quantity C_E depends upon the tricalcium aluminate (C3A) content of the cement, which the user must obtain from a chemical analysis of the cement.

This model predicts the velocity R at which the degradation front advances through the concrete specimen. As the front advances into the specimen, 4SIGHT simply advances the external boundary conditions with the front. Since sulfate attack completely destroys the concrete in its wake, advancing the boundary conditions is equivalent to treating the destroyed concrete like soil. When a sufficient quantity of the upper surface of the roof has deteriorated due to sulfate attack, it will fail, at which time the calculation will terminate. This critical depth of sulfate attack before failure is a parameter that must be supplied by the user.

Leaching

Leaching of the calcium hydroxide is not modelled directly. Rather, leaching is incorporated into the program as an effect of maintaining chemical equilibrium. After each time increment, each computational element is brought to chemical equilibrium, the details of which are presented subsequently. If necessary, chemical equilibrium is achieved through dissolution or precipitation of available salts. Changes in the quantity of salts changes the porosity of the cement paste. This, in turn, changes the transport coefficients.

Cracks and joints

Although cracks are not a degradation mechanism, their effects must be incorporated. Since the roof will be supported, its bottom surface will be in tension, assuming simple support conditions at the boundaries. Therefore, it is expected that the bottom surface of the roof slab will contain cracks. The cracks are approximated by parallel plates, originating from, and perpendicular to, the bottom surface, and extending part way into the slab. Assuming Poisseuille flow[4], the permeability of two plates separated by a distance w is

$$k_p = \frac{w^2}{12} \tag{2}$$

If the permeability of the uncracked concrete is k_o, and the cracks are spaced a distance a apart, the bulk permeability is

$$k_b = k_o\left(\frac{a}{a+w}\right) + k_p\left(\frac{w}{a+w}\right) \tag{3}$$

The same analysis can be extended to concrete joints. Given the permeability of the joint material, k_j, the bulk permeability can again be calculated using Eqn. 3. An additional consideration is the lifetime of the joint, which must be specified by the user. Upon failure of the joint, its permeability may increase by ten orders of magnitude. At this point, the useful life of the concrete has ended and the calculation terminates.

The cracks/joints are implemented in the one-dimensional model by using averaged quantities. For example, consider cracks originating at the bottom of the slab, and extending one half the distance to the top of the slab. The permeability of the top half of the computational elements is that of the sound concrete. The permeability of the bottom half of the elements is calculated from Eqn. 3.

Advection-Diffusion

The flux, $\mathbf{j}$ of ions due to both diffusion and a volume averaged velocity, $\mathbf{u}$, is

$$\mathbf{j} = -D\nabla c + c\mathbf{u} \tag{4}$$

where c is the ion concentration, and D is the diffusion coefficient, or diffusivity. The time rate of change in concentration is the negative divergence of the flux [5]:

$$\frac{\partial c}{\partial t} = \nabla \cdot D\nabla c + c\,\nabla \cdot \mathbf{u} + \mathbf{u} \cdot \nabla c \tag{5}$$

Note that D may be a function of location due to leaching. Also, the term $\nabla \cdot \mathbf{u}$ may not be zero due to the changing porosity. The volume averaged velocity, $\mathbf{u}$ can be related to the Darcy velocity,

$$\mathbf{v} = -\frac{k}{\mu}\nabla p \tag{6}$$

through the relationship

$$\mathbf{v} = \phi\mathbf{u} \tag{7}$$

The above equations are combined and implemented in a one-dimensional finite difference algorithm with sufficiently short time steps to insure a stable solution. This system of equations can be used to propagate ions through the system given the material parameters of the concrete, the external ion concentrations, and the hydrostatic pressure.

Chemical Equilibrium

4SIGHT currently recognizes the ions H^+, Ca^{2+}, Na^+, K^+, OH^-, Cl^-, SO_4^{2-}, and CO_3^{2-}, and all the two-component salts that can be formed from these ions. After each time step, each computational element is brought to chemical equilibrium through a combination of solubility limits and charge conservation. For a salt composed of anion $A^{\alpha-}$ and cation $C^{\beta+}$, the concentration of each ion in a element containing the salt must satisfy the solubility product relationship. Additionally, for all the m anions and all the n cations present in a computational element, the program must insure charge conservation. A simultaneous solution to both constraints is achieved by precipitation or dissolution of available salts.

Physical Parameters

All of the concrete physical parameters (*e.g.*, diffusivity, permeability, *etc.*) are user-specified inputs to 4SIGHT. However, in cases where not all properties are available, missing quantities must be approximated using existing correlations. The physical properties must be established due to both hydration and leaching. The physical properties due to hydration are the initial conditions. However, as the porosity changes due to leaching, corrected values of the physical parameters are needed.

Hydration

The hydration of cement can be approximated by a reaction between tricalcium silicate (C3S) and water, forming a calcium silicate hydrate (CSH) and calcium hydroxide. After some period of hydration, the fraction of the initial C3S which has hydrated is the degree of hydration, α. The relation between these two properties and capillary porosity can be estimated from the water-cement ratio, $\frac{w}{c}$, and from stoichiometric relations for cement paste[7]:

$$\phi_o = 1 - \frac{1 + 1.31\alpha}{1 + 3.2\frac{w}{c}} \tag{8}$$

Although this equation was developed for C3S, the factor of 1.31 is not expected to vary by more than about 10% for typical cements [8].

The diffusivity can be related to either $\frac{w}{c}$ or ϕ using an empirical relation between D and $\frac{w}{c}$ for cement paste [9, 10]:

$$\log_{10} D_o = 6.0\frac{w}{c} - 13.84 \tag{9}$$

The dimension of D_o is m^2/sec. This equation is an estimate of the ultimate diffusivity of the concrete. It does not incorporate any degradation.

There is a universal relationship between D and ϕ [11] for cement pastes:

$$\vartheta = \frac{D}{D^f} = 0.001 + .07 * \phi^2 + 1.8(\phi - .18)^2 H(\phi - .18) \tag{10}$$

where D is the concrete diffusivity, D^f is the free ion diffusivity, and $H(x)$ is the Heaviside function. The quantity ϑ represents the ratio $\frac{D}{D^f}$ due to hydration and is a constant for all ions.

The above equations relate the diffusivity of cement paste to $\frac{w}{c}$ or porosity. A relationship is now needed for the relationship between cement paste diffusivity and concrete diffusivity. Experimental results of Luping and Nilsson[12] for cement paste and mortar suggest that their diffusivities are approximately equal. Additionally, numerical experiments of Garboczi, Schwartz, and Bentz[13] which investigate the effect of interfacial zone diffusivity upon bulk diffusivity suggest that the bulk diffusivity is approximately equal to the paste diffusivity.

Given $\frac{w}{c}$, the permeability of concrete can be approximated from the data in Hearn, et al.[14]:

$$\log_{10} k_o = 5.0\frac{w}{c} - 21 \tag{11}$$

the dimension of k is m^2.

Leaching

Once D and k have been established, changes due to leaching can be calculated from $\vartheta_o = \vartheta(\phi_o)$, where ϕ_o is the initial porosity due to hydration, calculated from Eqn 8. As the $Ca(OH)_2$ is leached, the porosity increases. Let the value of porosity after leaching be ϕ', and the diffusivity be D'. The ratio D'/D^f is not simply $\vartheta' = \vartheta(\phi')$ because as calcium hydroxide is leached from the paste the ratio D'/D^f does not follow Eqn. 10.

The NIST cement microstructural model [15] was used to determine the relation for D'/D_o upon leaching. Using this model, an empirical relation was developed to relate the formation factor of the leached pore structure to the undamaged pore structure:

$$\xi = \frac{D'}{D^f} = \vartheta_o + 5.0 * (\vartheta' - \vartheta_o) \tag{12}$$

Therefore, the ratio of the leached value of diffusivity, D', to the initial value D_o is

$$\frac{D'}{D_o} = \frac{\xi}{\vartheta_o} \tag{13}$$

The relative change in permeability can be calculated using ξ, the Katz-Thompson equation [16], and the fact that the critical pore diameter, d_c is proportional to ϑ [17]:

$$\frac{k'}{k_o} = \left(\frac{\xi}{\vartheta_o}\right)^3 \tag{14}$$

Validation

At present, the validation of `4SIGHT` has been limited to comparisons to analytic equations for nonreactive species being transported by either diffusion or Darcy flow. Validation of service lif prediction is hindered by the small number of published reports of service life experiments which include periodic measurements of material properties.

Distribution

The program `4SIGHT` has been precompiled for a personal computer using the MSDOS operating system, and can be obtained from

`ftp://titan.cbt.nist.gov/pub/4SIGHT`

Questions and comments can be addressed to `4SIGHT@titan.cbt.nist.gov`.

Summary

Upon reducing the problem of estimating the service performance of an LLW facility concrete vault to its most critical element, a reasonable estimate of service life can be attained by a one-dimensional model incorporating diffusion and advection. Reducing the problem to one of monitoring the transport of ions through the concrete and maintaining chemical equilibrium allows for the synergism of multiple degradation mechanisms.

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